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# Composite membranes for electrochemical hydrogen separation: models and materials

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Electrochemical permeation membranes for hydrogen separation operate by the oxidation of hydrogen to protons at one side (the anode), transport of protons and electrons or holes across the membrane, and reduction to a hydrogen species at the opposite side (the cathode). For single phase materials, the process is described as ambipolar diffusion [1]. The fact that many conventional proton conductors such as  $\text{BaCe}_{1-x}\text{Zr}_x\text{Y}_y\text{O}_{3-y/2}$  (BCZY) are oxides doped with lower valence elements is unhelpful to the development of single phase membranes, since the hydrogen-rich conditions in which these must operate favor n-type, rather than p-type, electronic conduction. Despite that, membranes made with Tm and Ru doped perovskite proton conductors have been reported to give moderate hydrogen fluxes [2, 3].

An alternative approach is to separate the roles of the protonic and electronic conductor by using a composite, for example  $\text{SrZrO}_3/\text{SrFeO}_3$  [4] or  $\text{BaCe}_{0.2}\text{Zr}_{0.7}\text{Y}_{0.1}\text{O}_3$  /  $\text{Sr}_{0.95}\text{Ti}_{0.9}\text{Nb}_{0.1}\text{O}_3$ . [5]. While relaxing the requirement for two types of conductivity to coexist in one system, this approach introduces issues of fabrication, chemical compatibility and phase percolation, as well as issues of possible interfacial impedance at hetero-phase junctions.

In this paper, percolation is treated via effective medium models for two-phase systems and the effective ambipolar conductivity of a two-phase composite is derived according to these models. Composite  $\text{H}_2$  permeation membranes reported in the literature are discussed with reference to these issues, and also the surface reaction rate, which dominates performance at low temperatures.

## References

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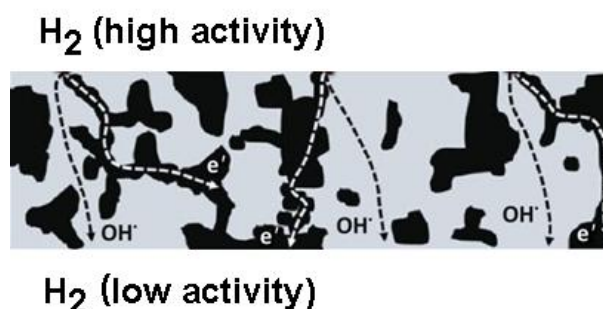


Fig. 1 Principle of operation of composite hydrogen permeation membrane.

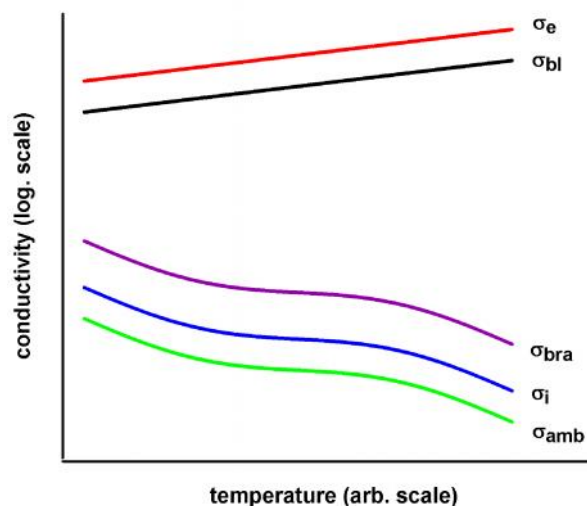


Fig. 2 Schematic conductivity plots for an electronic and an ionic conductor, and several conductivities that can be directly derived for a composite of 50/50 by volume:

- $\sigma_e$  electronic conductivity
- $\sigma_i$  ionic conductivity
- $\sigma_{bi}$  Bruggeman-Landauer model
- $\sigma_{bra}$  Bruggeman asymmetric model
- $\sigma_{amb}$  ambipolar conductivity (non interacting phases)